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Evidence of a first order like jump in equilibrium magnetization across the peak effect region in superconducting 2H-$NbSe_2$

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Abstract

We report magnetization hysteresis measurements in the peak effect region of a very clean single crystal of superconducting 2H-$NbSe_2$ at temperatures very close to $T_c(0)$. Through measurement of minor magnetization curves, we infer the equilibrium magnetization curve across the PE region. We observe a small first order like change in equilibrium magnetization across the peak effect. We relate this observation to the entropy change associated with the order-disorder transformation. We further note that the first order like change in the equilibrium magnetization observed in 2H-$NbSe_2$ is comparable to the change(s) observed in the equilibrium magnetization at the FLL melting transition in various high temperature superconductors.

The advent of high T_c superconductivity focused wide spread attention on pristine physics issue of melting of pure (pinning free) Abrikosov flux line lattice (FLL)[1]. There is now a growing and compelling evidence from magnetic[2-4], thermal[5] and structural studies[6,7] in a variety of superconductors, that the vortex solid to vortex liquid transformation is of first order (or weakly first order)[2-4]. The crucial input in favor of this has stemmed from the observation of a step increase in the equilibrium magnetization (ΔM_{eq}). At temperature values where FLL melting is anticipated, the Clausius- Clayperon equation,

$$L = T\ \Delta S = -T\ \Delta M_{eq}(dH_m/dT) \qquad (1)$$

provides the necessary connection between the latent heat L and ΔM_{eq} via the knowledge of the slope of the FLL melting transition (H_m, T) curve. However, the step increase in the equilibrium magnetization has been experimentally discerned only across limited (and different) $H-T$ regions in crystals of BiSrCaCuO [2] and YBaCuO [3]. In LaSrCuO system it could however be observed over a much wider (H, T) region[4]. Keeping in view that FLL melting is a generic phenomenon related to collapse of long range spatial order of FLL, its fingerprint(s) ought to be observable even in the mixed state of conventional low temperature superconductors across an appropriate (H, T) region. However, there are fewer reports of such claims in conventional low T_c compounds as compared to those in high T_c cuprates. FLL melting in conventional superconductors is difficult to investigate experimentally, because it is believed to occur very close to H_{c2}[1], where there could be additional complications arising from the fluctuations in the phase of the superconducting order parameter as well.

Two prerequisites[1] for facilitating the observation of FLL melting in a specimen of conventional superconductor are: (i) a large value of the Ginzburg number $G_i = (1/2)(kT/H_c^2\xi^3\epsilon)^2$; and (ii) the existence of appreciable pinning free region (where magnetization is reversible) below superconductor-normal phase boundary. Anisotropic hexagonal 2H-$NbSe_2$ system with $T_c(0)$ = 7.2K has a $G_i \sim 10^{-4}$[8]; this value lies in between those of high T_c cuprates (10^{-2}) and low T_c alloy superconductors (10^{-6}). This system is therefore considered appropriate to explore the phenomenon of FLL melting in the domain of conventional low T_c alloy superconductors. As stated above FLL melting is a pure system concept and in cuprate superconductors its fingerprint in reversible region of dc magnetization data is convenient to locate only in clean single crystal samples which are free of structural defects. The high purity single crystals of 2H-$NbSe_2$ system rank favorably amongst the most weakly pinned superconducting samples of all varieties of superconducting systems. The ratio J_c/J_0 (where J_c is the critical current density and J_0 is the depairing current density) in clean crystals of 2H-$NbSe_2$ is typically $\sim 10^{-6}$[8] and this value is orders of magnitude lower than those usually observed in conventional low T_c superconductors. However all high quality single crystals of 2H-$NbSe_2$ display the peak effect (PE) phenomenon[9], which is the anomalous increase in J_c close to $H_{c2}(T)$ where FLL melting is expected. Ever since the initial impetus injected by an explanation of PE phenomenon by Pippard[10] in terms of rapid collapse of rigidity of vortex

lattice, various (other) possible close connections between FLL melting and PE phenomenon have been provided from time to time via static and dynamical measurements[8] on vortex states as well as via theoretical treatments and simulation studies[11,12].

In isothermal dc magnetization measurements, the peak effect manifests as an anomalous increase in the magnetization hysteresis. It is understood that the width of the hysteresis loop at a given field H provides a measure of $J_c(H)$. Recent structural studies through small angle neutron experiments in single crystals of Nb[6] and μSR measurements in 2H-$NbSe_2$ [7] support the existence of an intimate relationship between the reduction in spatial order of FLL and the PE phenomenon. The entropy change associated with the collapse of long range order in the FLL therefore needs to be discerned from the change(s) in the equilibrium magnetization across the PE region. We report in this paper a successful outcome of our attempts to locate a step change in the equilibrium magnetization (ΔM_{eq}) across the peak effect region from the dc magnetization hysteresis data in a clean crystal of 2H-$NbSe_2$.

Isothermal magnetization data (across PE region) has been measured using a Quantum Design SQUID magnetometer with field parallel to c-axis of a clean 2H-$NbSe_2$ single crystal, at T = 6.85K and 6.95K respectively. The data at T=6.95K is recorded with a 2 cm scan. However at 6.85K, the magnetization hysteresis is comparable to the field inhomogeneity along the scan length thereby necessitating the use of half-scan technique [13] with an effective 4 cm scan. This particular crystal is very weakly pinned and the PE can be observed down to a field of 50 Oe in temperature dependent ac susceptibility (χ') experiment (see inset of Fig.1. for χ'(T) plot in H = 1kOe, however all data not shown here). The locus of peak temperature T_p vs H in it has the behavior (Fig.1) which lies in between those for single crystals A and B of 2H-$NbSe_2$ studied by us earlier (see Fig.4 of Ref [9]). In isothermal dc magnetization hysteresis data (see Figs 2(a) and 2(b)), the PE region has been identified with the anomalous increase in the hysteresis just below H_{c2} values. The peak fields H_p at 6.85K and 6.95K are consistent with $T_p(H)$ curve shown in Fig.1. It may be noted that the forward and reverse legs of the hysteresis envelope in the PE region are significantly asymmetric. Further, the field value H_{pl}^+ at which anomalous increase in the diamagnetic response commences on the forward magnetization curve differs significantly from the field $H_{pl}^-(< H_{pl}^+)$ where PE ends on the reverse leg.

In clean single crystals of high T_c superconductors, the dc magnetization is reversible over a wide range of fields prior to H_{c2} and any modulation (step

change, inflection points etc.) in equilibrium magnetization can therefore be identified distinctly. However, when the magnetization is irreversible, M_{eq} values are usually obtained[14] as,

$$M_{eq}(H) = [M^{+}(H) + M^{-}(H)]/2, \qquad (2)$$

where M^{+} and M^{-} are the magnetization values measured in ascending and descending field cycles. Each of these values comprises contributions from shielding currents set up in the sample in addition to the equilibrium magnetization. An implicit assumption in this relation is that the critical current density J_c at a given H remains the same on ascending and descending fields. In other words J_c is independent of magnetic history of the vortex state. In recent years transport[15], dc magnetization and ac susceptibility[16] studies have revealed that J_c in weakly pinned superconducting samples (which show PE phenomena) could strongly depend on their thermomagnetic history. While studying the effect of thermomagnetic history on transport critical currents in a crystal of Niobium which showed PE, Steingart et al [17] had noted the inequality,

$$J_c^{FC}(H) > J_c(H^{-}) > J_c(H^{+}), \qquad (3)$$

for fields below H_p (see Fig.2a for identification of peak field H_p). $J_c^{FC}(H)$ is the critical current density in field cooled (FC) state and $J_c(H^{-})$ and $J_c(H^{+})$ are the critical current densities measured in decreasing and increasing fields respectively. Originally, Steingart et al[17] had surmised that the vortex state in the field cooled state is most strongly pinned as each of the vortex lines attempts to conform to maximum number of pinning sites as the flux lines nucleate below H_{c2}. In view of the above inequality, Eq.(2) cannot be used for obtaining M_{eq} for $H < H_p$ and a suitable alternative has to be found so that the pair of magnetization values in Eq.2 correspond to the same value of $J_c(H)$. However, for $H > H_p$, where J_c is observed to be independent of magnetic history, M_{eq} could be obtained using Eq.2.

Within Larkin-Ovchinikov collective pinning description[18] (Larkin volume $V_c = R_c^2 L_c \propto J_c^{-2}$, where R_c and L_c are radial and longitudinal correlation lengths respectively) the inequality $J_c(H^{-}) > J_c(H^{+})$ implies that the extent of order in the vortex state generated in decreasing field (across PE region) from above H_{c2} is less than that in the vortex state created while increasing the field from zero value. In other words, the so called Larkin volume at a given field, over which FLL remains correlated, is larger on the increasing field cycle as compared to that on the decreasing field cycle. In an isothermal dc magnetization experiment, this inequality (which holds upto H_p) results in a hysteresis loop which is asymmetric (as shown in Fig.2a and

2(b)) with respect to the equilibrium magnetization because the contribution of the induced shielding current to the magnetization on the reverse leg would be larger in magnitude than that on the forward leg. Considering that the PE phenomena relates to reduction in spatial order of the vortex array, the observation $H_{pl}^{+} > H_{pl}^{-}$ implies that the fully disordered state, occurring at $H = H_p$ on the decreasing field cycle, does not fully heal back to the ordered state as field values are reduced. In fact the healing process continues at least down to $H = H_{pl}^{-}$. We believe that the existence of path dependence, i.e., $H_{pl}^{-} \neq H_{pl}^{+}$,is another manifestation of first order nature of the order-disorder transformation accompanying the PE phenomenon.

Roy and Chaddah[19] have proposed that the minor magnetization curves obtained by reversing the field from different values lying on the forward magnetization curve could be used to construct the requisite reverse leg of the hysteresis curve which is a symmetric counterpart of the forward magnetization curve[14]. Within Critical State model, such minor hysteresis curves merge into the reverse magnetization curve. However, in the PE region of $CeRu_2$, minor magnetization curves initiated from a field H lying between H_{pl} and H_p, saturate without merging into the usual descending envelope curve. They [19] have used the saturated value $M_{minor}^{-}(H)$ in Eq.1 instead of $M^{-}(H)$ to obtain $M_{eq}(H)$. We also show in Figs.2(a) and 2(b), the magnetization curves obtained while reversing the field from different values lying on the forward magnetization curve at 6.85K and 6.95K in our crystal of 2H-$NbSe_2$. We note that the minor magnetization curves originating from field values lying between H_{pl} and H_p do not reach upto the usual reverse magnetization curve, whereas those originating from $H > H_p$ merge with the reverse envelope curve which is consistent with the observations of in $CeRu_2$ [19] . In addition, we observe that for certain range of fields below H_{pl} also, the minor curves saturate without merging with reverse magnetization curve. We would like to assert that these minor curves are the symmetric counterparts (i.e., notionally correspond to same $J_c(H)$) of the forward magnetization curve and correspond to the more ordered FLL compared to that on the reverse curve. Following Ref [19], we use the saturated magnetization M_{minor}^{-} on the minor magnetization curve in Eq.1 to obtain M_{eq}. The more ordered FLL can alternatively be generated starting from a field cooled vortex lattice (for $H < H_{pl}$) and subjecting it to an increasing (FC-FOR) and decreasing (FC-REV) fields. Since the current density in FC state is much larger than those in the increasing and decreasing fields, the minor hysteresis loops initially overshoot [16] the envelope hysteresis curves as shown in Fig.3. FC-FOR

curve eventually merges into the forward magnetization curve, whereas the FC-REV curve merges into the minor magnetization curve initiated from the forward magnetization curve. It is pertinent to remark here that the minor magnetization curve (at 6.85 K) initiated from the forward curve merges into the usual reverse (obtained by reversing from H > H_{c2}) magnetization curve at fields sufficiently below H_{pl}. Also, it may be pointed out that if the field is increased from the (reverse) minor hysteresis curve, the magnetization readily merges into the usual forward envelope curve.

We understand the observed behavior of the minor magnetization curves within the LO collective pinning description [18] by the following argument. For a small decrement in field from H lying on the forward magnetization curve, the shielding currents merely reverse sign while the magnitude is maintained same as that on the forward magnetization curve. The size of Larkin domains or the extent of FLL correlations essentially remain unaltered. For the same field value H, on the reverse magnetization curve, the sign of the induced currents are same as those on the minor magnetization curve but are of larger magnitude. As stated earlier, a locus of the $M^-_{minor}(H)$ values obtained at different fields (see dotted curve in Fig.2a) appears reasonably symmetric with respect to the usual forward magnetization curve. Thus, if we use M^-_{minor} instead of M^- in Eqn.1, we obtain the M_{eq} as shown in the inset of Fig.2. The step increase in M_{eq} across the PE region can therefore be easily identified (see ΔM_{eq} as marked in the inset of Fig 2(a)). The tiny peak like modulation in M_{eq} values between H_{pl} and H_p is reminiscent of similar behavior across the PE region in some samples of $CeRu_2$ as reported in Ref. 19. It has been argued that the change in M_{eq} at H_{pl} in the case of $CeRu_2$ is an imprint of a first order onset of formation of a new superconducting phase with enhanced pinning. However, we attribute this ΔM_{eq} to the onset of amorphisation of FLL as a consequence of thermal softening of its elastic modulii across the PE region. Latent heat across the PE region in 2H-$NbSe_2$ can be estimated by substituting the value of dH_{pl}/dT ($\approx dH_p/dT$)$\approx$-5$\times 10^3$ Oe/K (Fig.1) along with ΔM_{eq} = 380mOe and 230 mOe at 6.95K and 6.85K, respectively. Our values of ΔM_{eq} and L compare favorably with similar estimates across FLL melting transition in cuprate superconductors[2-4] and across the PE region in $CeRu_2$.

To conclude, we have presented an estimate of step change in equilibrium magnetization (ΔM_{eq}) extracted from dc magnetization hysteresis across the PE region at 6.85 K and 6.95K at H_p 1.7 kOe and 1.0 kOe, respectively in a single crystal of 2H-$NbSe_2$ for H_{dc} parallel to c. From a variety of detailed

transport measurements in pure crystals of 2H-$NbSe_2$ which elucidated the dynamics of driven vortex matter prior to and across the PE region, it had been argued that [**?**] the vortex matter at peak position of PE region is in a *pinned liquid* state. Two recent structural studies via μ SR experiments [17] in this system have established that the spatial order of FLL undergoes a sudden change at the onset of PE in temperature dependent scans in 500 $< H <$ 200 Oe. Though PE region extends over 200 Oe in Fig.2(a) and 2(b), the peak effect phenomenon implies a sharp transformation in the state of vortex matter. The transition width of PE in temperature dependent ac susceptibility measurements (at fixed H) is smaller than the width of the normal to superconducting transition in zero field [9]. Thus, we believe that our estimate of Δ M_{eq} across PE reliably determines the latent heat associated with the occurrence of order to disorder transformation across PE. We find that the estimated value of ΔM_{eq} in 2H-$NbSe_2$ is of the same order as those observed earlier at FLL melting transition in crystals of cuprate superconductors [2-4]. We also note that our values of ΔM_{eq} in 2H-$NbSe_2$ also compare favorably with those reported across the PE region in some samples of superconducting $CeRu_2$[19].

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Figure 1: A comparison of Peak Effect curve (locus of T_p vs H) in 2H-$NbSe_2$ crystals used in the present study with those in the crystal A and B studied in Ref[9]. The dotted lines identify PE curves in crystals in A and B, whereas the filled circle data points in the present sample corresponds to peak temperatures T_p(H) as determined from ac susceptibility measurements shown in the inset. The T_p values in different crystals have been normalized to the respective $T_c(0)$ values. For the sake of completeness of the magnetic phase diagram the H_{c1}(T) and H_{c2}(T) curves have also been included in the main panel.

Figure 2(a): Isothermal magnetization hysteresis data for H$\|$c across the Peak Effect region in 2H- $NbSe_2$ at 6.85 K. $H_{pl}{}^+$ and $H_{pl}{}^-$ identify the field values at which PE notionally commences and terminates along the usual forward and reverse hysteresis paths. H_p identifies the peak field of PE. It also includes the minor hysteresis curves generated by reversing the fields from H = 1600 Oe and 1640 Oe and 1740 Oe lying on the forward hysteresis leg. The minor curves initiated from H = 1600 Oe (< H_{pl}) and 1640 Oe (H_{pl} < H < H_p) do not reach upto the reverse envelope curve, instead they lie just above the forward envelope curve. The dotted curve passing through the saturated values of $M_{minor}{}^-$ (see text or Ref.19) sketches the new reverse envelope curve which appears more symmetric with respect to the forward envelope curve as compared to the usually experimentally measured reverse envelope curve by decreasing the field from above H_{c2}. The equilibrium magnetization values prior to PE (i.e., for H < H_{pl}) lie in between the forward envelope curve and new reverse envelop curve, whereas the equilibrium value above the PE (H > H_{irr}) can be readily identified with the (path independent) measured (magnetization value. The inset shows the step change in equilibrium magnetization (ΔM_{eq}) across the PE region. Two straight line have been drawn in the inset to guide the eye about the occurrence of ΔM_{eq} across the PE region.

Figure 2(b): Isothermal magnetization hysteresis data for H_{dc} $\|$ c across PE region in 2H- $NbSe_2$ at 6.95 K. The identity of different symbols in this figure is the same as described in the caption of Fig.2(a). The inset shows the step change in equilibrium magnetization ΔM_{eq} across the PE region.

Figure 3: Minor magnetization curves generated by decreasing (REV) or

increasing (FOR) the field from field cooled magnetization value at H = 1300 (< H_{pl}) at 6.85 K. The FC - REV and FC - FOR curves initially overshoot the respective envelope curves thereby showing that $J_c{}^{FC}$ is much larger than the J_c values along the envelope curves. It is to be noted that FC-FOR curve readily merges into the forward envelope curve whereas the FC-REV curve drops down to values which lie significantly below the usually measured reverse envelope. In fact FC-REV curve appears to lie very close to the forward envelope curve, thereby implying very low values of J_c (for H < H_{pl}) for vortex states on the forward envelope curve.

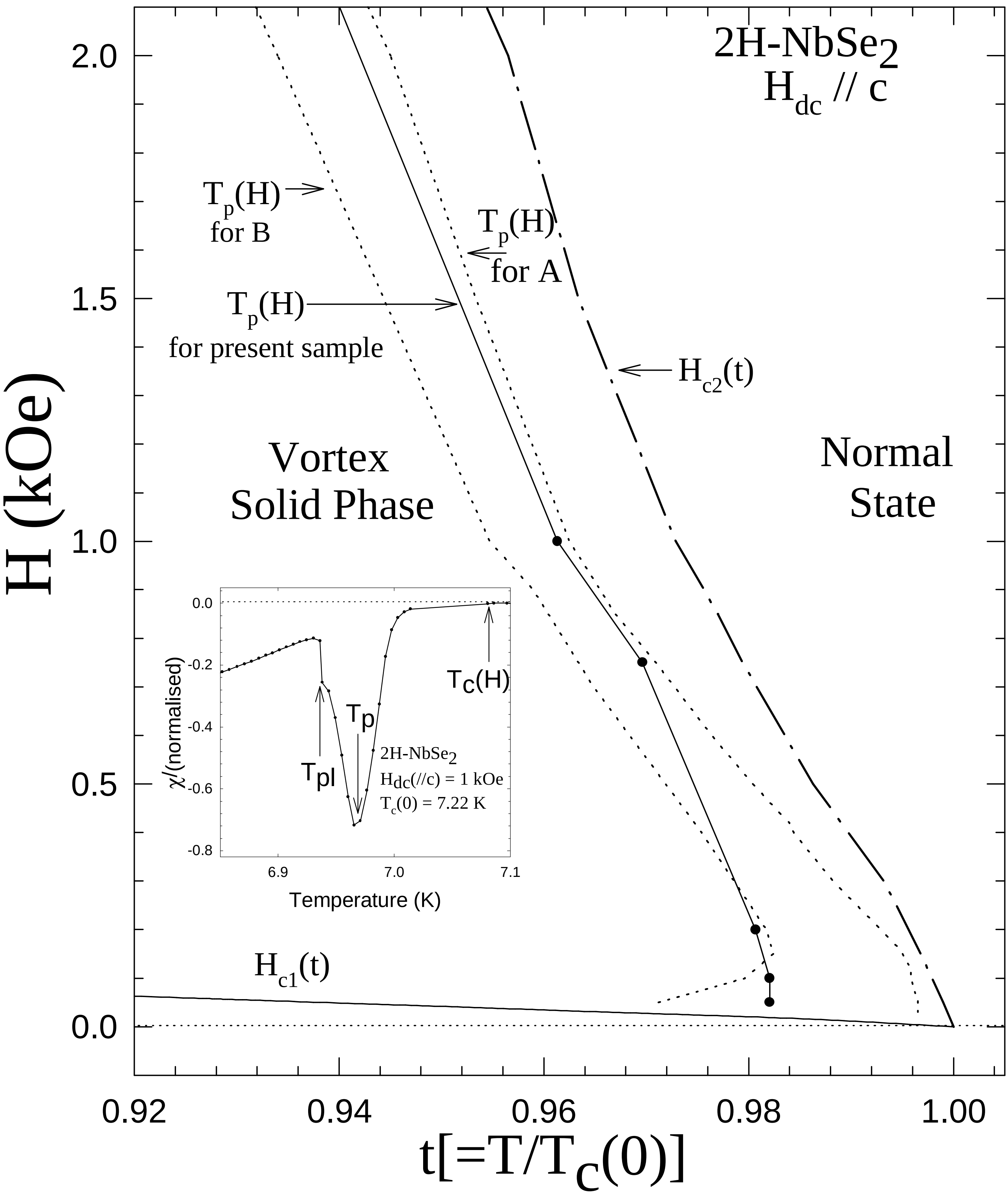


Fig.1 (Ravikumar et al)

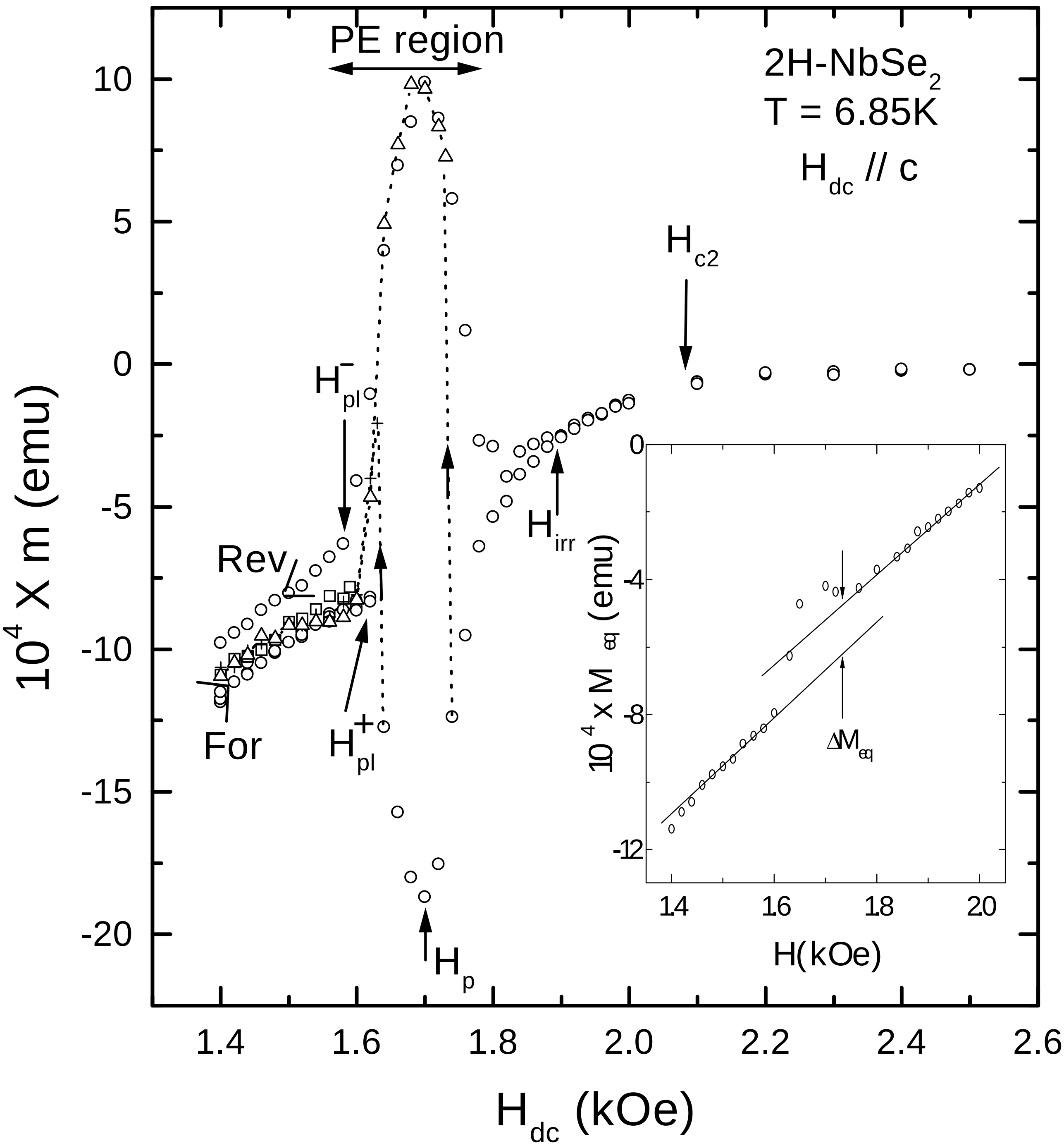
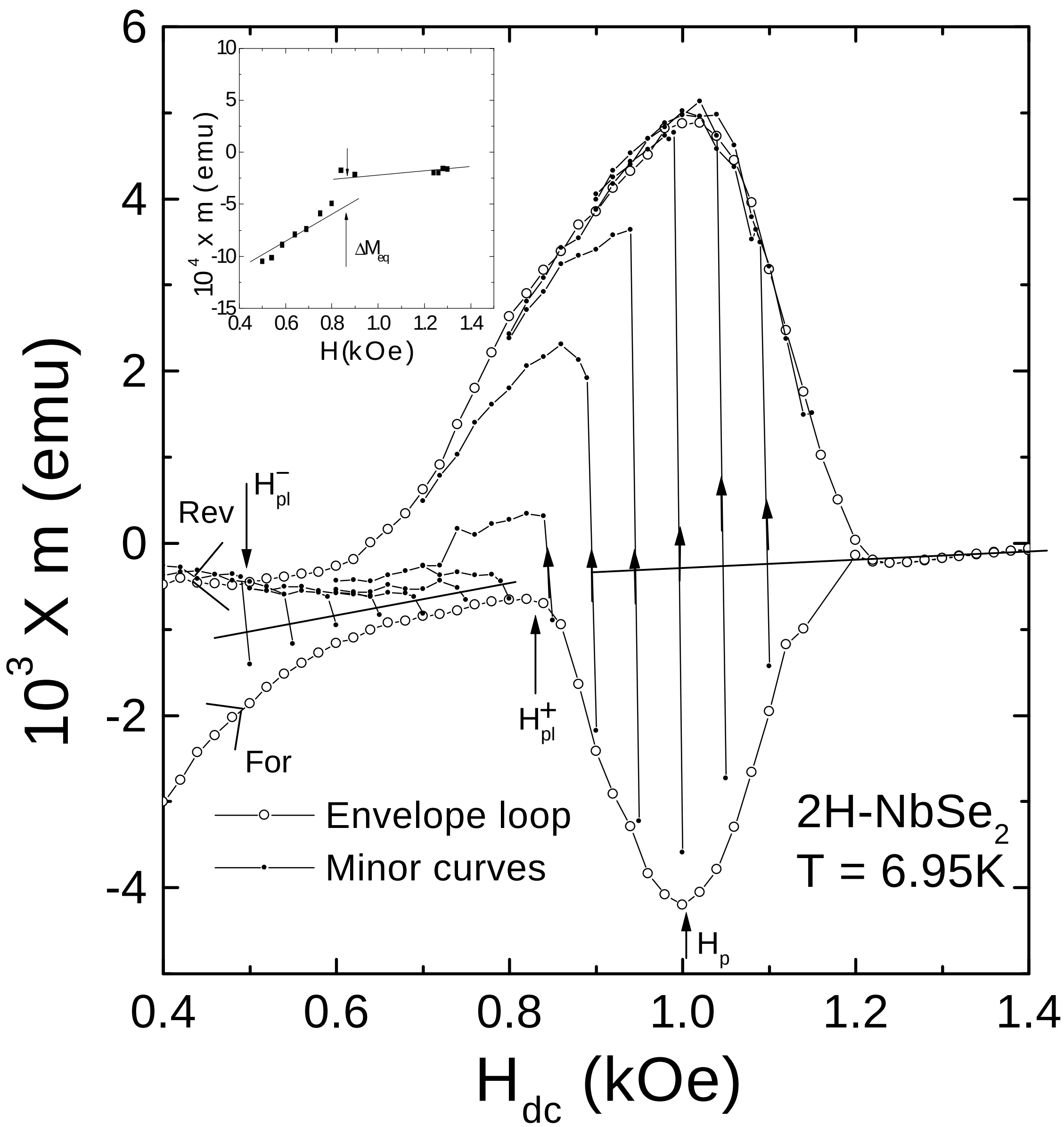


Fig.2(a) (Ravikumar et al)



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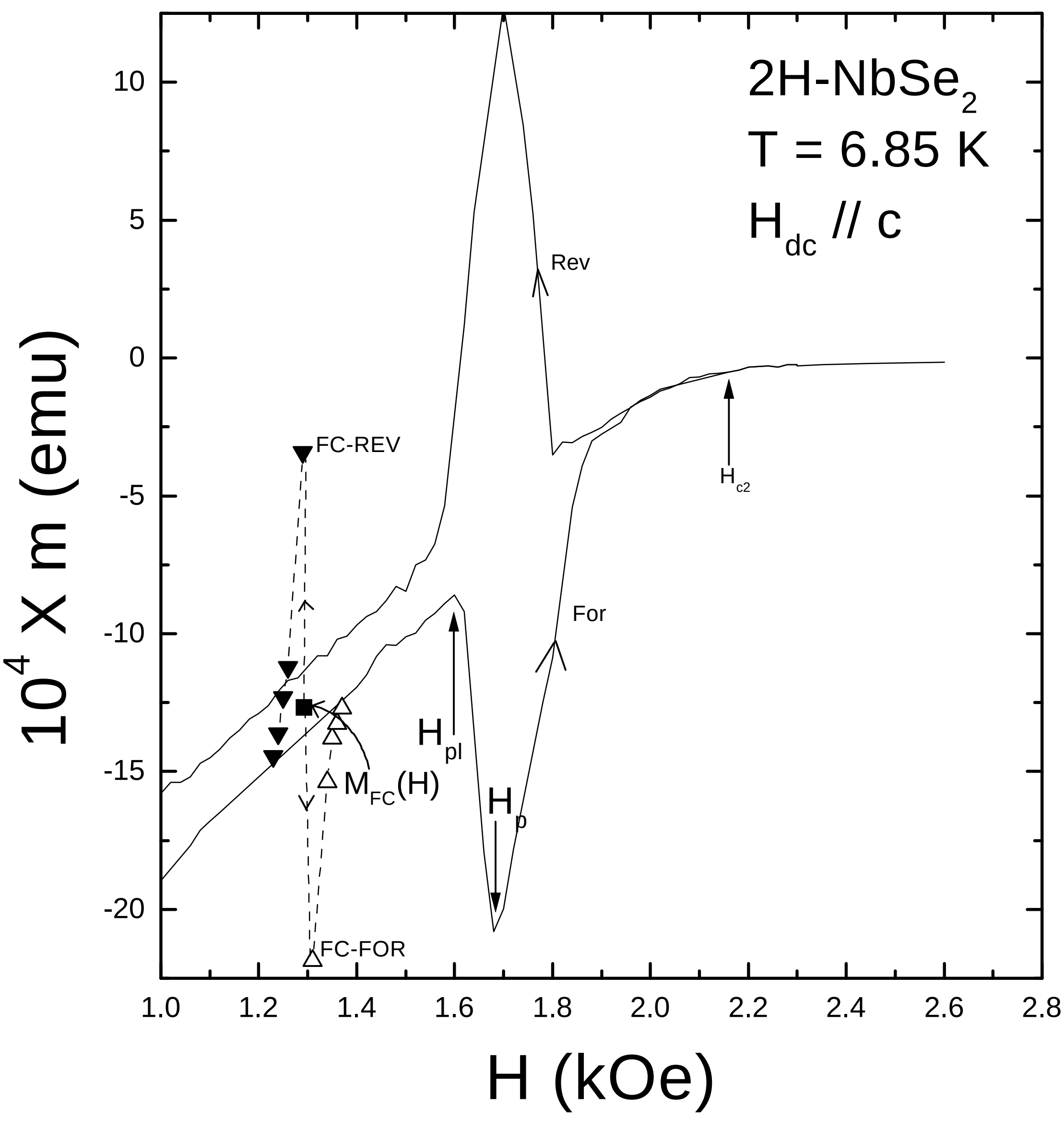

Fig.3 (Ravikumar et al)